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Absorption of SO_2 and NO through an integrative process with a cost-effective aqueous oxidant☆

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Summary Due to the substantial influence on the environment and health, there has been an intensifying environmental concern on sulfur dioxide (SO_2) and nitrogen oxide (NO) discharge to the atmosphere all over the world. Flue gas of coal-fired power stations is major leading source for the emission of SO_2 and NO. Cost-effective and balanced technologies for the reduction of these pollutants from flue gas have become increasingly essential at the present time. The preliminary experimental attributes were retrieved in a batch scale agitating bubbling reactor for instantaneous absorption of SO_2 and NO using aqueous oxidant. The prime conditions for instantaneous absorption examined were including absorbent concentration, reaction temperature, and pH in the fabricated flue gas system. Removal efficiencies of 99% for SO_2 and 85% for NO were acquired, respectively under absorbent concentration 5 g/100 ml, reaction temperature 313 K, and pH 5.6.

Introduction

Pollution prevention is an important task in current era throughout the world of industrial development. The significant air pollutants sulfur dioxide (SO_2) and nitrogen oxides (NO_x) emitted into atmosphere due to fossil fuels combustion leads the contamination impact on environment such as acid rain, photochemical smog, hazardous effects to ecosystem and so on. Previously several individual treatments have been carried out for removal of toxic gases separately. But multi gas removal of SO_2 and NO is an emerging trend in consideration of cost end optimistic results. Major contributions were carried in wet and dry basis. Wet gas removal was a feasible approach in these large scale operations. There are several effective absorbents tested for NO and SO_2 absorption such as ClO_2 (Jin et al., 2006),

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$FeSO_4$ (Zhang et al., 2010), Fe(II) EDTA (Wang et al., 2007), Ferrate (VI) (Zhao et al., 2014), $KMnO_4$/NaOH (Chu et al., 2001a), M/$NaClO_2$ (Mondal et al., 2013), $NaCi_3$/NaOH (Zhao et al., 2010) $NaClO_2$ (Chien and Chu, 2000), $NaClO_2$/Urea (Wei et al., 2009), $NaClO_2$/NaOH (Chu et al., 2001b), NaClO (Mondal and Chelluboyana, 2013; Chelluboyana and Kumar, 2014; Raghunath and Mondal, 2016), NH_3 (Dong et al., 2012), Urea (Fang et al., 2011), Urea/$KMnO_4$ (Fang et al., 2013), UV/H_2O_2 (Liu et al., 2010), etc.

This paper is concerned about the instantaneous removal of SO_2 and NO with a complex absorbent calcium hypochlorite ($Ca(OCl)_2$) using agitating bubbling reactor. Use of calcium hypochlorite will improvise the oxidation rate to convert NO to NO_2 and further reaction lead to absorption of SO_2 and NO. In this process concentrations of SO_2 and NO were taken as similar to the desired concentrations of SO_2 and NO in flue gas emission. A simulated gas blend has been prepared during the process. This gas blend was subjected to absorption and the effective parameters such as absorbent concentration, operating temperature, initial pH, initial SO_2 concentration and initial NO concentration were varied to study the feasibility of $Ca(OCl)_2$ as absorbent for integrative absorption of SO_2 and NO parameters in a bubble column.

Experimental

The schematic of experimental setup was detailed in Fig. 1 including with three major sections such as a simulated flue gas preparation section, an agitating bubbling reactor and a flue gas analysis section. The volume of the bubbling reactor was 250 m^3. A desired concentration of flue gas was prepared by using gas cylinders 1 (N_2), 2 (SO_2 with balance N_2) and 3 (NO with balance N_2) with two stage pressure regulators in gas mixing and pressure releasing chamber 4. To measure the initial concentrations of the gas stream, the valves V_4, V_7, V_9 were opened valves V_5, V_8 and V_{10} were closed so that the gas stream was send to multi flue gas analyzer 9 named garboard 3800 P (Wuhan Cubic Optoelectronics Co. Ltd., Wuhan, China). During the absorption process the gas stream was send to absorption column 6 covered with temperature controller water bath 7 to maintain the constant temperature throughout the process. The mixing of the gas stream to the liquid stream can be done with help of mechanical agitator 5. The agitator helps in bubble breakage and that leads to fluent mixing of gas phase with liquid phase. The valves V_1, V_2 and V_3 were kept open throughout the process. The total gas flow rate in the process was measured with help of gas flow meter 10 and the gas stream was discharged into atmosphere. The gas stream was dried with help of silica column 8 before sending to analysis. The readings were taken for every 10 min during the process till the absorbent reaches the saturation point.

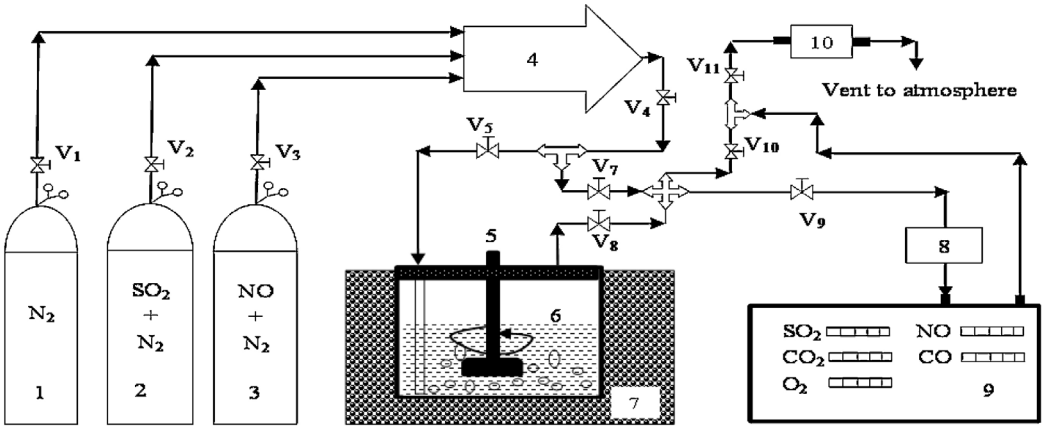
Results and discussion

Effect of reaction time

The change in absorption efficiency with respect to time has been determined in this process by analyzing the absorbed gas stream within regular time interval of 10 min keeping other parameters frozen. This has provided effectiveness of absorbent over SO_2 and NO absorption for a required contact time in the process. From the results it was observed that the optimum time for the absorption process for a 5 g/100 ml $Ca(OCl)_2$ was 2 h at initial SO_2 concentration of 6126.81 ppm, initial NO concentration 876.32 ppm, initial pH of 5.6 and temperature of 313 K.

Effect of absorbent concentration

The effect of absorbent concentration provides the capability and optimistic requirement of absorbent for simultaneous SO_2 and NO removal at required time. The effect of $Ca(OCl)_2$ at different concentration range 1—7 g/100 ml on simultaneous SO_2 and NO removal has been illustrated in Fig. 2. From the figure it was observed that the removal efficiency increased with increase in absorbent concentration till 5 g/100 ml of $Ca(OCl)_2$ and tends to



1. N_2 gas cylinder with two stage pressure regulator, 2. SO_2 + balance N_2 gas cylinder with two stage pressure regulator, 3. SO_2 + balance N_2 gas cylinder with two stage pressure regulator, 4. Gas mixing chamber, 5. Agitator blade, 6. Absorption reactor, 7. Water bath with temperature control, 8. Silica column, 9. Multi flue gas analyser, 10. Gas Flow meter

Figure 1 Schematic diagram of experimental set-up.

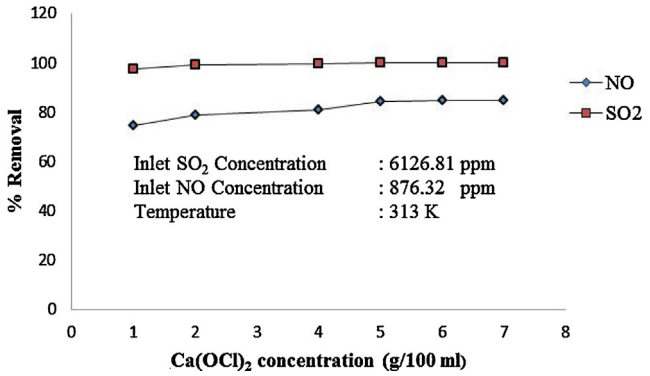


Figure 2 Effect of $Ca(OCl)_2$ concentration on simultaneous removal of NO and SO_2.

constant with increase in concentration. The maximum removal efficiency 85% was obtained for NO and 99% for SO_2 in this process. Form the data it was concluded that the optimal concentration given as 5 g/100 ml of $Ca(OCl)_2$. Initial dissolution of absorbent in water has been taken place and which lead to accumulate in terms of hypochlorite ions; these hypochlorite ions react with NO and convert if to NO_2 which proceed to formation of HNO_3, HNO_2 and simultaneously to H_2SO_4 due to dissolution of SO_2 in liquid phase. The efficiency of this process was effective and the absorbent can be considered in large scale operations.

Effect of absorbent temperature

Effect of reaction temperature on the process is a potential approach to study the change in removal efficiency with respect to the temperature of the absorbent. In this process the detailed illustration was given in Fig. 3 regarding the temperature change at a range of 293—323 K with respect to the removal efficiency of SO_2 and NO. This dissolved species of the absorbent in water react with simulated gas stream of SO_2 and NO in which operating temperature plays an effective role on gas—liquid reactions. The removal efficiency has been steadily increased with increase in temperature from 293 to 313 and then decreases. At the later stage, the decrease is due to the solubility of gas decreases with increase in temperature and the initial increase in removal efficiency is due to the reactive affinity of hypochlorite with NO in liquid phase. This is due to increase in presence vacant hypochlorite ions with increase in temperature due to solubility factor. The major intermediates formed during the

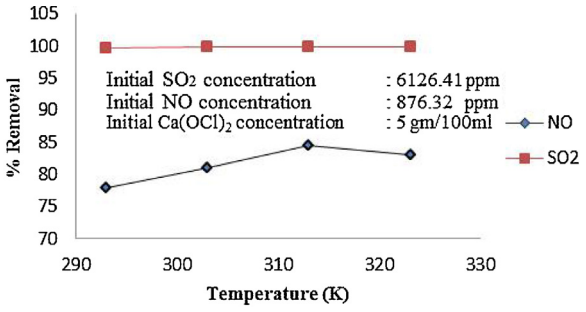


Figure 3 Effect of absorbent temperature on simultaneous removal of NO and SO_2.

absorption process initially were HClO, Cl^-, ClO^- ions in aqueous phase due to aqueous dissociation of $Ca(OCl)_2$ and additional absorption of SO_2 and NO by the agitation of solution were in the form of dissolved SO_3^{2-}, SO_4^{2-}, NO_2^-, NO_2^- and NO_3^- the main sulphur and nitrogen species in the liquid phase. From the data the optimal temperature feasible for this process was 313 K.

Effect of initial pH

The effect of initial pH of NO has been examined keeping 5 g/100 ml absorbent with reaction time 2 h, SO_2 concentration 6126.81 ppm; initial NO concentration 876.32 ppm and reaction temperature of 313 K. This study concludes the required optimum pH of the liquid containing $Ca(OCl)_2$ for simultaneous removal of SO_2 and NO. The Optimum of the process was pH obtained as 5.6.

Conclusions

Complex absorbent $Ca(OCl)_2$ has shown an effective removal efficiency of both SO_2 and NO of this integrative process. The optimum conditions for simultaneous removal were obtained at a process temperature of 313 K; absorbent concentration was 5 gram/100 ml and pH of 5.6 in the simulated flue gas absorption. Removal efficiencies of 99% for SO_2 and 85% for NO were accomplished, respectively under optimal experimental conditions.

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